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Diffuse pollution by persistent organic pollutants as measured in plastic pellets sampled from various beaches in Greece

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ABSTRACT

Plastic pellets found stranded on beaches are hydrophobic organic materials and thus, they are a favourable medium for persistent organic pollutants to absorb to. In the present study, plastic pellets are used to determine the diffuse pollution of selected Greek beaches. Samples of pellets were taken from these beaches and were analyzed for PCBs, DDTs, HCHs, and PAHs. The observed differences among pellets from various sampling sites are related to the pollution occurring at each site. Plastic pellets collected in Saronikos Gulf beaches demonstrate much higher pollutant loading than the ones collected in a remote island or close to an agricultural area. Based on data collected in this study and the International Pellet Watch program, pollution in Saronikos Gulf, Greece, is comparable to other heavily industrialized places of the world. The present study demonstrates the potential of pellet watch to be utilized as a detailed-scale monitoring tool within a single country.

1. Introduction

Plastic pellets, mainly polyethylene (PE) and polypropylene (PP), are widely distributed in the beaches all over the world (Ogata et al., 2009). These plastic pellets are unintentionally released to the marine environment during manufacturing and transportation. The program International Pellet Watch has resulted in maps for the global distribution of persistent organic pollutants (POPs) in coastal waters (Ogata et al., 2009). These maps demonstrate that using beached plastic pellets as passive sampling media it is possible to identify pollution hot spots in a global map. Plastic pellets were collected by different researchers throughout the world. After being sorted and sampled using the appropriate statistics, selected pellets were extracted with solvents that were then analyzed for certain groups of POPs.

In our previous paper (Ogata et al., 2009), we applied the pellet watch on global scale and spatial patterns of the concentrations of POPs were revealed. Concentrations of polychlorinated biphenyls (PCBs) were higher in the coasts of USA, Japan, and western European countries whereas minimal in tropical Asia. On the other hand, organochlorine pesticides DDTs showed the maximum concentrations in the samples from Vietnam. These large-scale spatial patterns were ascribed to differences in the chemical usage among countries. However, inputs of chemicals to coastal waters are spatially variable within individual countries and consequently chemical pollution varies in small-scales as well. To make effective controls on chemical pollution, understanding the detailed distribution of the pollutants and identifying the hot spots within different countries are essential. The pellet watch approach has potential for such identification. Endo et al. (2005) suggested the nation-scale spatial variation in PCB concentrations in the pellets from Japanese coast. However, at that time, sorting and analytical protocol has been under construction and no concrete results were obtained. So far, no evaluation of the pellet watch approach has been published for the detailed scale investigation of coastal pollution by using the currently established protocol.

In the present study, the pellet watch approach is applied for a smaller scale area. Greece is a small country with long shore line, and closed seas (e.g. Aegean Sea, Ionian Sea, etc.) that belong to a bigger closed sea, the Mediterranean Sea. More specifically, the objectives of the present study are (a) to explore the ability of plastic pellet sampling method to determine differences in the seawater pollution of beaches found in a small country like Greece and (b) to determine the POPs pollution status of seawater in four different beaches in Greece.

Plastic pellets have been identified in Greek beaches in a previous study (Karapanagioti and Klontza, 2007). Clean and polluted beaches should be included to determine whether the plastic pellets as monitoring media can be used for smaller scales. The Saronikos Gulf was monitored as the polluted site since it is one of the hotspots of the Mediterranean Sea for PCBs, DDTs, and HCHs pollutants (Galanopoulou et al., 2005; Gómez-Gutierrez et al., 2007). Pollution was attributed to the charge of the area through

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the central Athens sewage outfall. At the Saronikos Gulf both Loutropyrgos beach and Aegena island were sampled (Fig. 1, Table 1). An open sea beach (Vatera, Lesvos island) (Fig. 1, Table 1) and Kato Achaia beach in the Gulf of Patras were selected as non-polluted sites (Fig. 1, Table 1).

2. Materials and methods

Plastic pellets were sampled from four different locations in Greece (Table 1). In sandy beaches, plastic pellets can be found on the top of the sand. Samples were taken from the high tide line as well as from the berm or the upper part of the beach. Plastic pellets were then separated from the sand and stored in aluminum foil at 4 °C. They were shipped to Japan via airmail for further analysis.

The pellets were sorted by near-infrared spectrometer (Plascan-WTM OPT Research Inc., Tokyo, Japan) into PE, PP, and other polymers. Among the PE samples, yellowing pellets were selected by naked eye by comparison with the reference pellets whose yellowness had been determined by a Handy Colorimeter (NR-3000, Nippon Denshoku Ind., Ltd., Japan). These yellowing PE pellets were then subjected to the chemical analysis since these were proven to accumulate higher concentrations of polychlorinated biphenyls (PCBs) (Endo et al., 2005; Ogata et al., 2009). To consider piece-to-piece variation, 5 pools (each pool consists of 10 pieces of pellets) were analyzed for each location and the median value is reported in this study. Pellets were extracted with hexane by maceration. The extracts were purified by silica gel column chromatography. Each time pools from a certain beach were analyzed, a blank solvent sample was also analyzed along with these 5 sample pellet pools.

Nineteen major PCB congeners (CB-8, CB-18, CB-28, CB-52, CB-44, CB-66, CB-101, CB-110, CB-118, CB-105, CB-149, CB-153, CB-138, CB-158, CB-128, CB-187, CB-180, CB-170, CB-206),18 major polyaromatic hydrocarbons (PAHs) and dichlorodiphenyldichloroethylene (DDE) were measured by gas chromatograph equipped with ion-trap mass spectrometer on MS/MS mode. Dichlorodiphenyltrichloroethane (DDT), dichlorodiphenyldichloroethane (DDD) and hexachlorocyclohexanes (HCHs) were measured by GC equipped with an electron capture detector (GC-ECD).

3. Results and discussion

Plastic pellets collected in Greek beaches are mainly made from polyethylene (Table 2) followed by polypropylene and other different materials.

Concentrations of most PCBs in the beached pellets are higher (almost an order of magnitude) than in the blank solvent samples (Table 3). In all beaches, PCB-138 has the highest concentrations when compared to other PCB congeners. Total concentrations of PCBs in the pellets from the Saronikos Gulf (Loutropyrgos and Aegena) are two orders of magnitude higher than in the pellets from the non-polluted beaches (Figs. 1 and 2). PCB profile for Kato Achaia is different from the other beaches (In Fig. 2a, compare peaks for congeners PCB-149 and 153 and PCB-180 and 170). However, the PCB profiles for the most polluted beaches and the non-polluted Vatera beach are similar. These results are expected since the area north of Saronikos Gulf is highly industrialized. In addition, Saronikos and Patras Gulfs are very enclosed seas. Kato Achaia beach receives the outfall of the industrial zone of Patras which is the nearest larger city. However, the industrial activity is mainly focused on food processing and thus, the discharge of PCBs is not expected. Based on the Poseidon weather system, Vatera during July 2009 (no data for July 2008 are available) received waves coming only from north–east or north west directions. This suggests that the pollution in Vatera is not related to Saronikos Gulf

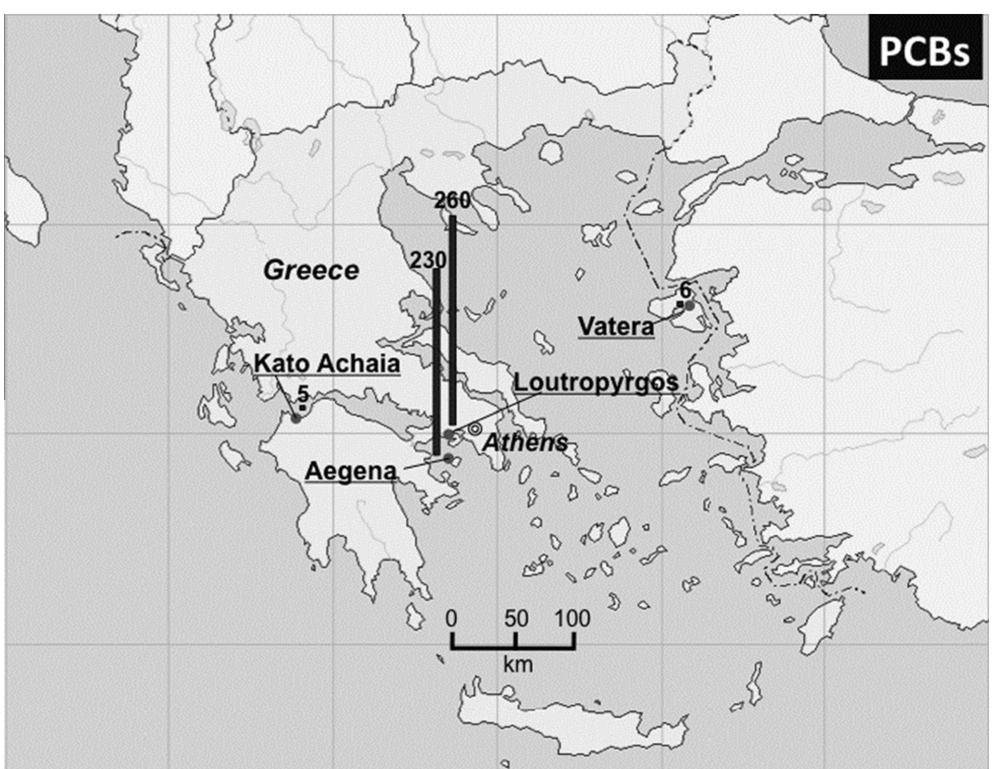

Fig. 1. Median concentrations of $\sum$13 PCBs (ng/g-pellet) in beached plastic pellets. $\sum$13 PCBs = sum of concentrations of CB# 66, 101, 110, 149, 118, 105, 153, 138, 128, 187, 180, 170 and 206.

Table 1
Sampling site details.

Sampling site	Location	Location	Sampling date
Kato Achaia	Kato Achaia beach, Gulf of Patras	38°09′ 18″N 21°33′ 00″E	March 2008
Vatera	Vatera beach, Lesvos island	39°01′ 07″N 26°11′ 27″E	August 2008
Loutropyrgos	Loutropyrgos beach, Gulf of Elefsis, Saronikos Gulf	38°01′31″N 23°28′28″E	October 2008
Aegena	Aegena island, Saronikos Gulf	37°43′ 30″N 23°31′ 53″E	October 2008

Table 2
Plastic pellet material (%).

	PE	PP	Other
Kato Achaia	54	32	14
Vatera	87	13	0
Loutropyrgos	90	10	0
Aegena	78	22	0

Table 3
Median concentration of PCBs in pellets (pg/g-pellet) from the four sampling sites (M) and corresponding blank solvent sample measurements (B).

	Kato Achaia		Vatera		Loutropyrgos		Aegena	
	B	M	B	M	B	M	B	M
CB-8	–	–	1	63	1	99	1	30
CB-18	–	–	25	100	37	600	23	630
CB-28	–	–	19	160	27	11,000	16	680
CB-52	–	–	15	63	9	4200	5	610
CB-44	–	–	23	63	31	4100	19	270
CB-66	6	200	12	43	14	10,000	9	620
CB-101	17	360	14	190	5	11,000	3	4400
CB-110	12	400	21	200	7	22,000	4	13,000
CB-118	7	230	18	180	11	10,000	7	4800
CB-105	10	90	7	56	6	4400	4	1900
CB-149	7	590	7	310	8	28,000	5	27,000
CB-153	7	610	11	940	7	37,000	4	32,000
CB-138	3	1300	17	1300	4	55,000	3	70,000
CB-158	–	–	0	0	8	4400	5	5000
CB-128	9	210	10	110	8	5400	5	7000
CB-187	1	450	12	400	10	21,000	6	13,000
CB-180	4	320	7	1000	14	37,000	8	32,000
CB-170	1	360	17	720	10	22,000	6	20,000
CB-206	5	6	8	10	4	180	3	57
TOTAL	–	–	240	6000	220	290,000	130	230,000
(CB66-206)	90	5000	160	5500	110	270,000	66	230,000

pollution although the PCB pattern is similar. Pollution is probably originating from industrial activity around Instabul or Northern Greece.

DDT is the original compound and DDE and DDD are the degradation products formed after the introduction of DDT to the environment. Aerobic conditions lead to the formation of DDE whereas anaerobic conditions lead to the formation of DDD. Ratios of DDE/DDT and DDD/DDT much lower than unit suggest a recent application. On Kato Achaia beach, DDE presents higher values than DDT (Table 4) suggesting a legacy pollution of DDTs applied in the past. When DDTs present higher concentrations than PCBs it is suggested that older pollution, mainly from agricultural and less from industrial sources, is prevalent. The Peiros River that outfalls in this beach receives the runoffs from an agricultural area and also the wastewater treatment plant outfall from a local municipality; in the past, this river used to receive the untreated wastewater from both an industrial site and the local municipality. On Vatera beach, DDTs concentration in the beached pellets was similar with that in the blank solvent sample (Table 4) suggesting that agriculture is not intensive in the area and there are no other sources for DDT pollution. On the Saronikos Gulf beaches, DDTs are 1–2 orders of magnitude higher than those in the blank solvent samples, but DDE/DDT ratios are not small enough to suggest recent pollution. This possibly suggests the use of these pesticides in industrial applications since limited agricultural land use can be found in the surrounding areas of the gulf. This is also collaborated by the high ratios of PCBs to DDTs.

Several decades ago, fish was sampled for PCBs and DDTs in the Saronikos Gulf (Satsmadjis and Gabrielides, 1979). These results can be used as a baseline contamination by POPs in the 1970s. At that time, fish sampled close to Loutropyrgos presented lower ratio of PCBs to DDTs (median = 1.7) than nowadays (median = 15). Also, PCBs and DDTs found in fish sampled close to Loutropyrgos were 19 and 11 times higher when compared to Aegena, respectively whereas concentrations for both sites are similar in the present days (median = 1.04 and 0.43 times higher and lower, respectively). This suggests that industrial pollution spreads throughout the Saronikos Gulf whereas agricultural pollution shrinks.

In all sampled beaches, HCH concentrations are close (lower than 3 times higher) to procedural blanks (Table 5). These pesticides are found at lower concentrations than DDTs, which is in agreement with the study conducted for various waterbird species from Greece (Sakellarides et al., 2006). In a recent study that analyzed sediment from the port of Piraeus, Saronikos Gulf, similar trends are observed (Galanopoulou et al., 2005). Sum of HCHs are 1–2 orders of magnitude lower than the sum of DDTs. DDE/DDT and DDD/DDT ratios are not low enough to suggest a recent application. However, in this study (Galanopoulou et al., 2005), the sum of DDTs concentrations in the sediments is higher than the sum of 11 PCBs concentrations. As in the present study, this suggests that DDTs could originate from other applications in addition to agriculture since the port of Piraeus is surrounded by industrial activity.

No significant PAH concentrations are detected in the pellet samples from Kato Achaia and Vatera non-polluted beaches (Table 6). In the Saronikos Gulf beaches, PAH concentrations found in the beached pellets are 1–2 orders of magnitude higher than those in the blank solvent samples. Chrysene presents the highest concentrations in the sampled pellets. Actually, in Loutropyrgos beach, chrysene represents the one fifth of the total PAHs concentration. Other abundant PAHs include fluoranthene, phenanthrene, 2-methyl phenanthrene, anthracene, and indeno[1, 2, 3-cd]pyrene.

The sum of 9 PAHs concentrations measured in pellets in this study can be compared with the sum of the same PAHs measured in seawater and different parts of mussels sampled from the same beaches in Saronikos Gulf (Valavanidis et al., 2008; Fig. 3). The PAHs concentrations in the pellets are about 2 times lower than in mussels. However, when comparing sites, almost the same ratio value is observed for pellets and mussel gills (3.4 and 3.2, respectively); on Loutropyrgos beach PAH concentrations are about 3 times higher than in Aegena. Mussel mantles present a similar ratio value of 2.4. Water also showed higher concentration for Loutropyrgos than Aegena but the ratio calculated was higher (8.7) than for pellets and mussel gills or mantles. This higher ratio in water can be explained by the fact that within the PAH group there are compounds with varying hydrophobicities and thus, they will have a different distribution in the different media (e.g. water

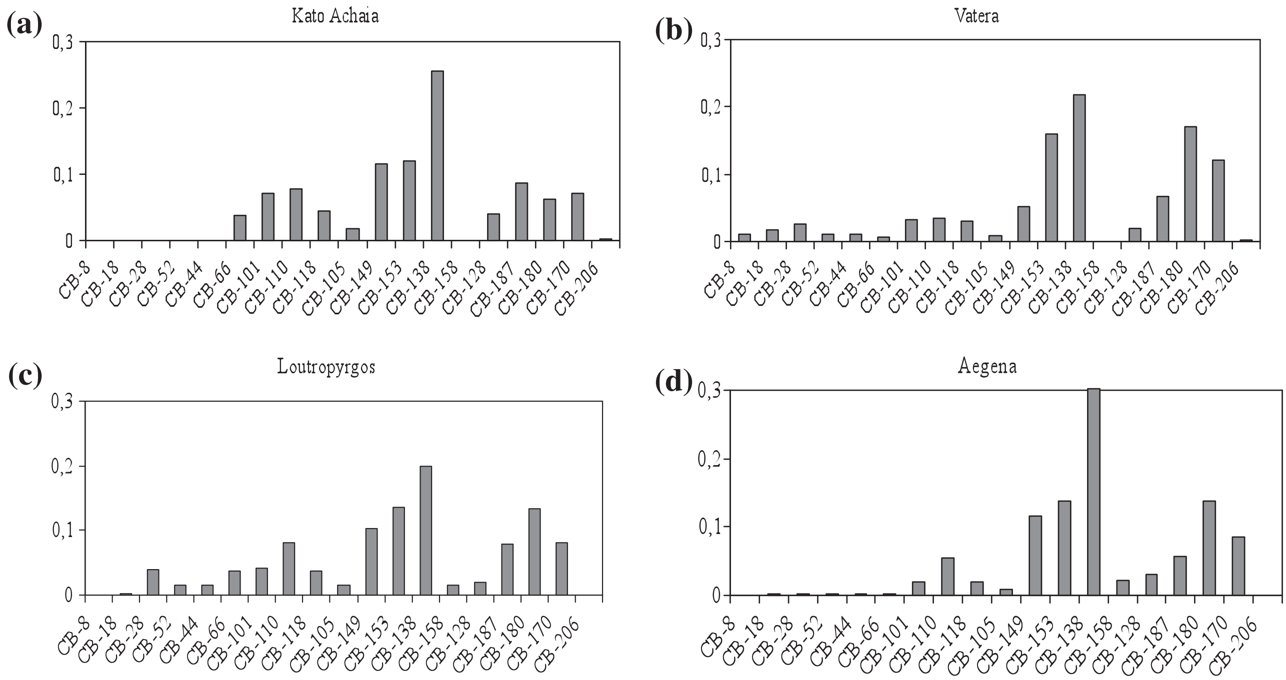

Fig. 2. PCB profiles ($C_{congener}/C_{sum}$) for all four sampling sites.

Table 4
Median concentration of DDTs in pellets (ng/g-pellet) from the four sampling sites (M) and corresponding blank solvent sample measurements (B).

	Kato Achaia		Vatera		Loutropyrgos		Aegena	
	B	M	B	M	B	M	B	M
DDE	0.02	10	0.22	0.25	0.11	6.0	0.07	15
DDT	0.2	1.2	0.34	0.56	0.61	9.9	0.37	25
DDD	0.2	0.8	0.40	0.32	0.67	2.2	0.41	2.1
TOTAL	0.5	12	0.97	1.1	1.4	18	0.84	42
DDE/DDT	–	8.7	–	–	–	0.60	–	0.61
DDD/DDT	–	0.72	–	–	–	0.22	–	0.08

Table 5
Median concentration of HCHs in pellets (ng/g-pellet) from the four sampling sites (M) and corresponding blank solvent sample measurements (B).

	Kato Achaia		Vatera		Loutropyrgos		Aegena	
	B	M	B	M	B	M	B	M
α-HCH	0.10	0.28	0.00	0.19	0.60	0.27	0.37	0.23
β-HCH	0.32	0.51	0.26	0.45	0.14	1.4	0.08	0.62
γ-HCH	0.09	0.16	1.5	1.3	0.96	1.3	0.59	0.75
σ-HCH	0.06	0.09	0.25	0.27	0.02	0.51	0.01	1.41
Total	0.57	1.05	2.1	2.2	1.7	3.5	1.1	3.0

versus pellets or mussel gills and mantles). Different compounds partition with varying degrees in plastic pellets from the water phase (Ahn et al., 2005). It is obvious that plastic pellet concentrations are generally representative of the water phase concentrations but describe even better the pollution within the body of aqueous organisms such as the gills of mussels.

If the degree of sorption for PAHs into the plastic pellets attained in the field is compared to the sorption attained in the laboratory at equilibrium (Karapanagioti and Klontza, 2008) or to theoretical calculations (Karapanagioti et al., 2010), it is observed that this degree is different between the two sites. This difference is not uncommon for environmental samples and also considering that the measurements found in the literature were performed at different times and by different research teams (Valavanidis et al., 2008). PAHs with molecular weights 178 and 202 reach around 1% of equilibrium in Loutropyrgos and 0.4% in Aegena (Karapanagioti et al., 2010). PAHs with molecular weight of 252 reach 10^{-4} of equilibrium in Loutropyrgos and 10^{-5} in Aegena (Karapanagioti et al., 2010). Much lower concentrations of PAHs in the pellets than those expected based on the aqueous concentrations can also be partly ascribed to photodegradation of PAHs because they are always exposed to sunlight. For these particular sites, these observations mean that in order to consider the one sampling site more or less polluted than the other, the ratios of the concentrations of lower molecular weight PAHs in the pellets between sites has to be higher than 4 and the same ratio for the higher molecular weight PAHs has to be higher than one order of magnitude (Karapanagioti et al., 2010). In any case, considering both the results from the pellets and also from the seawater literature data, it can be concluded that Loutropyrgos is more polluted by PAHs than Aegena.

In all cases, the ratio of phenanthrene concentration to anthracene concentration is lower than 10 suggesting that PAHs in these sites are of pyrolytic origin (Budzinski et al., 1997). This pollution is due to intense shipping activities, industrial and vehicular exhaust emissions (Valavanidis et al., 2008). The same conclusions related to pollution sources can be obtained analyzing beach pellets. Thus, plastic pellet watch approach seems promising even in a smaller scale as well as global.

Based on the present study and compared to other sampling beaches on earth studied by the International Pellet Watch program, Kato Achaia and Vatera beaches demonstrate low pollution as previously considered, whereas the beaches in Saronikos Gulf (Loutropyrgos and Aegena) demonstrate really high pollution levels (Ogata et al., 2009). The Saronikos Gulf presents: (a) the highest pollution based on PCBs concentrations in Mediterranean Sea, and lower concentrations than Japan and East Asia, North Sea, and California beaches, (b) DDTs concentrations lower than East Asia and California beaches, and (c) HCHs concentrations similar to other places on earth.

Table 6
Median concentration of PAHs in pellets (ng/g-pellet) from the four sampling sites (M) and corresponding blank solvent sample measurements (B).

	Kato Achaia		Vatera		Loutropyrgos		Aegena	
	B	M	B	M	B	M	B	M
Phenanthrene	7	6	27	33	3	34[a]	2.1	21[a]
Anthracene	4	2	6.7	2.8	1	36[a]	0.8	2.7[a]
3-Methyl phenanthrene	9	5	0.6	2.3	1	17	0.4	8.4
2-Methyl phenanthrene	11	7	0.7	2.8	1	32	0.2	17
9-Methyl phenanthrene	3	1	0.6	2.0	1	19	0.6	9.5
1-Methyl phenanthrene	5	5	4.2	2.1	1	17	0.3	7.6
Fluoranthene	4	7	15	11	3	86[a]	2.5	9.8[a]
Pyrene	5	8	9.9	6.4	2	36[a]	1.4	5.5[a]
Benz[a]anthracene	0	0	0.0	0.0	0	3	0.0	1.0
Chrysene	9	23	12	8.0	4	100	3.0	41
Benzo[b]fluoranthene	18	19	3.1	4.6	3	5[a]	1.7	2.3[a]
Benzo fluoranthenes	10	0	5.6	1.1	1	24	0.9	9.8
Benzo[e]pyrene	11	19	8.6	4.0	4	7	2.5	9.3
Benzo[a]pyrene	6	17	7.4	4.2	3	3[a]	2.4	2.8[a]
Perylene	5	19	7.2	2.9	3	2	2.3	4.0
Indeno[1,2,3-cd]pyrene	12	0	0.2	2.1	0	15	0.0	18
Benzo[ghi]perylene	6	12	12	12	2	3	1.5	2.2
Coronene	8	7	1.1	1.8	2	2	0.8	1.4
Total PAHs	130	160	120	100	35	500	23	180

[a] Used in Karapanagioti et al. (2010) to calculate percentages of sorption equilibrium attained within pellets.

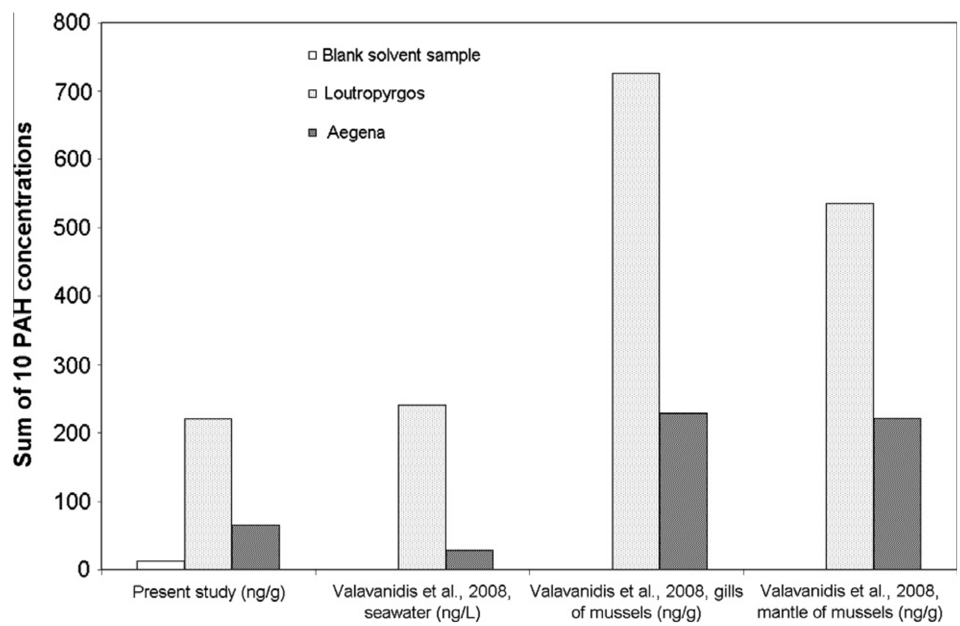

Fig. 3. Sum of 9* PAH concentrations in different phases sampled from the Saronikos Gulf. Ratios of 9* PAH concentrations in Loutropyrgos compared to Aegena are: (a) from the present study (sampling 2008): 3.4 in pellets, and (b) calculated from data published in Valavanidis et al. (2008) and sampling performed between 2005 and 2006: 8.7 in seawater, 3.2 in gills of mussels, and 2.4 in mantle of mussels (9* PAHs: phenanthrene, anthracene, fluoranthene, pyrene, benz[a]anthracene, benzo[b]fluoranthene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, benzo[ghi]perylene).

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